

Comment on “Precision measurement of the Casimir-Lifshitz force in a fluid”

B. Geyer¹, G. L. Klimchitskaya^{1,2}, U. Mohideen³ and V. M. Mostepanenko,^{1,4}

¹*Center of Theoretical Studies and Institute for Theoretical Physics,
Leipzig University, D-04009, Leipzig, Germany*

²*North-West Technical University, Millionnaya Street 5, St.Petersburg, 191065, Russia*

³*Department of Physics and Astronomy,
University of California, Riverside, California 92521, USA*

⁴*Noncommercial Partnership “Scientific Instruments”,
Tverskaya Street 11, Moscow, 103905, Russia*

Abstract

Recently J. N. Munday and F. Capasso [Phys. Rev. A **75**, 060102(R) (2007)] claimed that they have performed a precision measurement of the Casimir force between a sphere and a plate coated with Au, both immersed in ethanol. The measurement results were claimed to be consistent with the Lifshitz theory. We demonstrate that the calculation of the Casimir force between the smooth surfaces of the test bodies following the authors prescription has a discrepancy up to 25% with respect to the authors result. We also show that the attractive electrostatic force only due to the surface potential differences was underestimated by a factor of 590. The resulting disagreement with the experimental data might be partially decreased by the effect of the charge double layer screening which was not taken into account. All this leads to the conclusion that the results of this experiment are in fact uncertain.

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In Ref. [1] the measurements of the attractive Casimir force between an Au-coated sphere and a plate immersed in ethanol using an atomic force microscope are presented. The obtained experimental data are compared with the Lifshitz theory taking into account the frequency dependence of the dielectric functions of Au and ethanol, and the correction due to surface roughness. Consistency of the obtained data with Lifshitz's theory is claimed, although at separation distances below 50 nm, disagreement with theory has been observed which increases with the decrease of separation. The performed experiment is interesting as a test of the Lifshitz theory for applications to three-layer systems and as a step towards the observation of the Casimir repulsion predicted in such systems. However, as we show below, the theoretical computations of the Casimir force between smooth Au surfaces separated by ethanol done according to the prescription provided by Ref. [1] leads to a discrepancy up to 25% with respect to the results of Ref. [1]. This increases the observed discrepancy between experiment and theory. A second drawback is that the effect of the residual potential difference between the sphere and the plate was calculated incorrectly and significantly underestimated by a factor of 590. Finally, the interaction between the double layer formed in liquids, which would decrease the electrostatic force, was not taken into account without any justification. We show that the resulting electrostatic force is of the same order of magnitude as the Casimir force to be measured. All these make the interpretation of this experiment uncertain.

The sphere-plate Casimir attraction across ethanol was calculated using the Lifshitz formula (2) of Ref. [1]. The dielectric function of Au along the imaginary frequency axis was found using the Kramers-Kronig relation (Eq. (3) in [1]) and optical data from the optical properties handbook [2]. The dielectric function of ethanol was taken from [3] (it was previously used in [4]). The computational results are presented in Fig. 3 of Ref. [1] as a dotted line for perfectly smooth surfaces of a sphere and a plate.

We have repeated the same computation using the same algorithm for the determination of the dielectric function of Au along the imaginary frequency axis, and the dielectric function of ethanol, as it is explicitly presented in [4]. Our results are shown by the solid line in Fig. 1. In the same figure the computational results obtained in Ref. [1] are presented as dots. As is seen in Fig. 1, the solid line significantly deviates from the dots over the measurement range from $d = 34$ nm to $d = 90$ nm. At $d = 34$ and 40 nm this deviation reaches 25% and 21%, respectively, of the force magnitude shown by the dots. It decreases to 6.5% at $d = 90$ nm

(at larger separations it is impossible to retrieve the force values in Fig. 3 of Ref. [1]).

Our precise computation of the dielectric function of Au along the imaginary frequency axis on the basis of the optical handbook data [2] can be found in Refs. [5, 6]. The computations of the Casimir force between Au surfaces using this function have been compared with independent computations made by other groups [7, 8], and agreement at the level from 0.1% to 0.2% was achieved. Bearing in mind that the dielectric function of ethanol is defined analytically [4], there is no problem in the inclusion of an intermediate liquid layer. This permits us to conclude that the dotted line in Fig. 1 is in disagreement with the practically coincident computational results of three above mentioned independent groups shown as the solid line. As is claimed in [1] with a reference to [9], the calculated force between gold surfaces in vacuum can vary by as much as 5% due to the variation in the optical properties that occur for different samples. This point is however irrelevant to the difference between the solid and dashed lines in Fig. 1, because if the data set reportedly used in Ref. [1] is employed (tabulated data [2] for Au and analytic function for ethanol), the results must be in agreement.

We admit that the roughness correction in Ref. [1] is computed correctly (the roughness profiles are not provided in [1] and, thus, the respective computations cannot be independently repeated), we arrive at the conclusion that the disagreement between experiment and theory in Ref. [1] is larger than indicated. For example, at $d = 40$ nm the deviation between experiment and theory in [1] including surface roughness is equal to $\approx 63\%$ of the measured force magnitude. If one takes into account, however, that the force magnitude between smooth surfaces at $d = 40$ nm is 28.5 pN larger than indicated in [1] (i.e., 163.5 pN instead of approximately 135 pN) the deviation between experiment and theory reaches $\approx 76\%$.

We emphasize that the computational results in Fig. 1, indicated as dots, can be reproduced if one uses at all imaginary frequencies the Drude dielectric function for both sphere and plate materials,

$$\varepsilon_1 = \varepsilon_2 = 1 + \frac{\omega_p^2}{\xi(\xi + \gamma)}, \quad (1)$$

instead of the optical handbook data, where $\omega_p = 9.0$ eV is the plasma frequency and $\gamma = 0.035$ eV is the relaxation frequency. It has been known [7] that the use of the Drude function instead of the optical tabulated data of [2] leads to large underestimation of the force magnitude at short separations. Recall that the text of Ref. [1] claims that the Kramers-

Kronig relation and the dielectric function of Au from the handbook [2] were used. The Drude dielectric function is not mentioned in Ref. [1].

The next point of this Comment is the effect of the residual potential difference between the sphere and the plate. According to Ref. [1], “the contact potential between the plate and the sphere in air is $V_0 = 130$ mV, as determined by varying the bias voltage on the sphere while keeping the plate grounded.” Reference [1] calculates the resulting electrostatic force in ethanol using the formula

$$F_{electrostatic} = -\frac{\pi R \varepsilon_0 V_0^2}{\varepsilon_{ethanol} d} = -10.326 \text{ pN at } d = 40 \text{ nm} \quad (2)$$

and considers the obtained value negligible compared to the Casimir force equal to -260 pN.

This calculation is in error. In the process of measurements both the sphere and the plate in Ref. [1] were kept grounded. Theoretically, for grounded bodies the potential difference V_0 , as determined in the air, remains the same in the presence of ethanol. In this case, the ground plays the role of a voltage source and compensates the screening effect of the polarization of ethanol. However, Eq. (2) used in Ref. [1] is derived under the condition that at a fixed separation d the total charge Q on each plate of the capacitor is fixed and is the same in air as well as in the dielectric fluid (Ref. [10], page 30, problem 1.7,a). It is not applicable to the case of grounded bodies where the charge cannot be controlled. In this situation, contrary to the authors statement on p.2 of Ref. [1], the electrostatic force between the sphere and the plate is not reduced but rather increases when the intervening material is a dielectric and not vacuum (see, e.g., Ref. [10], page 30, problem 1.7,b). In electrodynamics the residual potential difference remains equal to V_0 whether or not there is an intervening material while the capacitance in the presence of ethanol is increased by $\varepsilon_{ethanol}$. Using the proximity force approximation, this leads to a different equation for the residual electrostatic force between the sphere and the plate

$$\begin{aligned} F_{electrostatic} &= 2\pi R E(d) = -\frac{\pi R \varepsilon_0 \varepsilon_{ethanol} V_0^2}{d} \\ &= -6097 \text{ pN at } d = 40 \text{ nm}. \end{aligned} \quad (3)$$

Here, $E(d) = -CV_0^2/(2S)$ is the energy per unit area of a parallel-plate capacitor of area S with fixed potential difference V_0 between the plates and $C = \varepsilon_0 \varepsilon_{ethanol} S/d$ is the capacitance in the presence of ethanol.

The analysis of real experiments should be guided by the above theoretical considerations. Experimentally, there is no direct control either on Q or V_0 in liquids. While the variations

of Q can be arbitrary, the variations of V_0 can be considered as small for grounded surfaces. The value of V_0 is in fact controlled by the difference in the chemical potentials of the two metals. For this reason the electrostatic force can be estimated at constant V_0 rather than at constant Q . This is still an estimation. In fact it can happen that even V_0 can have small variations both depending on the separation and on the introduction of ethanol. Thus, in the experiment under consideration the residual electric force due to V_0 is in fact much larger than the Casimir force and must be taken into account in the comparison of experiment with theory.

The seeming contradiction between the large residual electric force (3) and the relatively small measured forces, as reported in Fig. 3 of Ref. [1], might be explained by an important effect that was not taken into account in Ref. [1] without any justification. This is the effect of the double layer formed near the surfaces of the plate and the sphere in ionic solution [11, 12]. Just like in a plasma, the ions from the salt impurities (even parts per million are sufficient while parts per thousand in ethanol are possible [13, 14]) will screen the surfaces leading to a decrease of the magnitude of the electrostatic force. This screening has been experimentally investigated in Refs. [12, 15] for a tip of an atomic force microscope above a plate immersed in different liquids. The electrostatic force itself can be attractive (always for conductive surfaces which are grounded, as in Ref. [1]) or repulsive (e.g., for two insulating surfaces). Thus, in either case the double layer effect acts to decrease the strength of the attraction or repulsion. The total electrostatic force between the grounded surfaces including the effect of the double layer in its simplest approximation takes the form (for a general case see Refs. [11, 12])

$$F_{electrostatic} = -Ae^{-d/\lambda}, \quad (4)$$

where λ is Debye length and A is a coefficient depending on the parameters of the system. For a large Au sphere of about $20\text{ }\mu\text{m}$ radius and a plate immersed in ethanol the corresponding A can be of several hundred pN. This is achieved for ion concentration between $10^{-4} - 10^{-5}$ moles per liter for ethanol used in the experiment of Ref. [1], much less than the solubility limit for a typical impurity such as NaCl.

It is easily seen that the electrostatic force (4) with account of the double layer is of the same order of magnitude as the expected Casimir force and may contribute significantly to the observed experimental data shown as circles in Fig. 3 of Ref. [1]. An accurate account of the effect of the net electrostatic force for the experimental configuration used is necessary

before any comparison between experiment and theory can be made.

To conclude, Ref. [1] contains an interesting attempt to measure the Casimir force in a three-layer system. The obtained results, however, cannot be considered as a “precision measurement” consistent with Lifshitz’s theory, as is claimed by the authors. In the above, we have demonstrated large errors in the calculation of the Casimir force and, most notably, of the residual electrostatic force. We have also indicated that the important effect of Debye screening due to the double layer which could partially compensate the large residual electrostatic force was not taken into account. In our opinion, this effect is a realistic possibility to explain the large disagreement between the experimental data in [1] and the magnitudes of the residual electrostatic forces between the grounded surfaces calculated on the basis of classical electrodynamics. The above remarks demonstrate that the results of Ref. [1] concerning the observation of the Casimir force in a three-layer system are uncertain.

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- [1] J. N. Munday and F. Capasso, Phys. Rev. A **75**, 060102(R) (2007).
 - [2] *Handbook of Optical Constants of Solids*, ed. E. D. Palik (Academic, New York, 1985).
 - [3] L. Bergström, Adv. Colloid Interface Sci. **70**, 125 (1997).
 - [4] J. N. Munday, D. Iannuzzi, Yu. Barash, and F. Capasso, Phys. Rev. A **71**, 042102 (2005).
 - [5] A. O. Caride, G. L. Klimchitskaya, V. M. Mostepanenko, and S. I. Zanette, Phys. Rev. A **71**, 042901 (2005).
 - [6] R. S. Decca, D. López, E. Fischbach, G. L. Klimchitskaya, D. E. Krause, and V. M. Mostepanenko, Ann. Phys. (N.Y.) **318**, 37 (2005).
 - [7] A. Lambrecht and S. Reynaud, Eur. Phys. J. D **8**, 309 (2000).
 - [8] J. S. Høye, I. Brevik, J. B. Aarseth, and K. A. Milton, J. Phys. A: Mat. Gen. **39**, 6031 (2006).
 - [9] I. Pirozhenko, A. Lambrecht, and V. B. Svetovoy, New J. Phys. **8**, 238 (2006).
 - [10] B. I. Bleaney and B. Bleaney, *Electricity and Magnetism*, vol. 1, 3rd ed. (Oxford University

Press, Oxford, 1989).

- [11] V. A. Parsegian and V. Gingell, *Biophys. J.* **12**, 1192 (1972).
- [12] J. Israelachvili, *Intermolecular and Surface Forces* (Academic Press, San Diego, 1992).
- [13] S. P. Pinho and E. A. Macedo, *Fluid Phase Equilibria* **116**, 209 (1996).
- [14] S. P. Pinho and E. A. Macedo, *J. Chem. Eng. Data* **50**, 29 (2005).
- [15] A. L. Weisenhorn, P. Maivald, H.-J. Butt, and P. K. Hansma, *Phys. Rev. B* **45**, 11226 (1992).

Figures

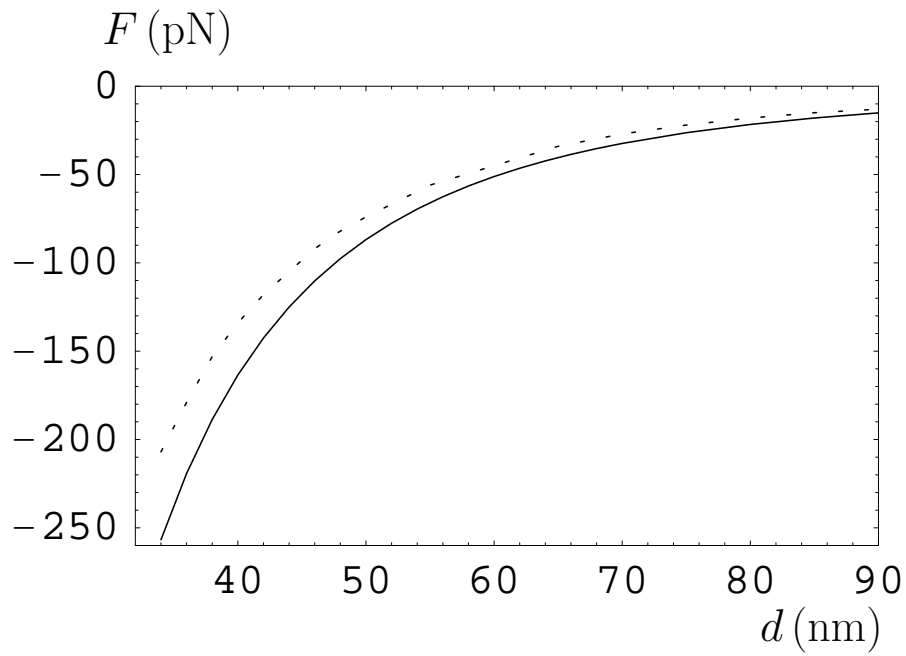


FIG. 1: The Casimir force between perfectly smooth gold surfaces of a sphere and a plate immersed in ethanol calculated in Ref. [1] (dotted line) and in this Comment (solid line) using the same procedure as a function of separation (see text for details).